



Landau-Devonshire Parameters for the Tunable Paraelectric Material $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$

by Virginia Miller and Frank Crowne

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14. ABSTRACT <p>Measured values of the nonlinear electrical response of the perovskite ferroelectric compound $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$ are used to deduce the thermodynamic Landau-Devonshire parameters of the material, which are found to differ strongly from those of the parent material BaTiO_3, despite the small amount of added Sc and Ta. The compound is found to cubic, with a low transition temperature of -60°C. An explanation for this behavior is advanced based on quenching of electrostriction.</p>					
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1. Introduction

The electronic properties of ferroelectrics make them attractive candidates for incorporation into various kinds of electronic devices. Because the performance of such devices in a given circuit environment depends in a highly specific way on the response of the material to external perturbations such as electric fields, stresses, and temperature, it is important to provide the device designer with an appropriate set of parameters that characterize this response, which may or may not be easy to obtain. In contrast, when a chemist wishes to synthesize a new material, he needs a different set of responses, based on thermodynamics and chemistry. Thus, it is vital to connect these differing responses mathematically so that the chemist can translate data from the device designer into a form that allows him to vary the parameters in the proper way.

In this report we attempt such a connection for the specific example of a promising new ferroelectric material, $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$. This material, which was fabricated by a novel process developed by S. Tidrow and the first author of this report, exhibits a strong and nonlinear ferroelectric response, namely a dependence of its dielectric constant on an external bias voltage. Such behavior, which is referred to as tunability and is common to many ferroelectric materials, is an example of a property that can be exploited by device designers. However, because it is intimately related to a strong increase in its dielectric constant with temperature near the transition from ferroelectric to paraelectric, it is very temperature-sensitive, an undesirable feature for electronic device applications. Moreover, most procedures that decrease the temperature sensitivity also decrease the tunability, so that it is important to determine what steps can be taken to decouple these processes.

2. Thermodynamics of Ferroelectrics

The analysis used in this report is based on the macroscopic theory of ferroelectricity developed by L. D. Landau (1937) and A. F. Devonshire (1949), now referred to as mean-field theory, which postulates the existence of a free energy per unit volume of ferroelectric that is analytic at the Curie temperature T_C , i.e., possesses a power-series expansion around that temperature:

$$F = \frac{1}{2}a(T - T_C)D^2 + \frac{1}{4}bD^4 + \frac{1}{6}cD^6$$

Here $D = \epsilon_0 E_{ex} + P$ is the electric displacement, E_{ex} is an externally applied electric field, and P is the polarization, i.e., the dipole moment per unit volume, whose nonzero value in the absence of an external electric field defines the ferroelectric state (I).

In a “normal” material, the minimum value of this free energy is obtained when $D = 0$. The quantities a , b , c , and T_C are referred to as Landau-Devonshire parameters. They are all characteristic material properties, which the material scientist can vary by modifying the synthesis procedure, and are assumed to be temperature-independent (2).

In the presence of an external field, the displacement can be computed by minimizing this free energy with respect to D , yielding the polynomial expression

$$E_{ex} = a(T - T_C)D + bD^3 + cD^5 \quad (1)$$

As $E_{ex} \rightarrow 0$ this polynomial has only the trivial solution $D = P = 0$ when $T > T_C$, while for $T < T_C$ it has several nonzero solutions in addition to $D = 0$. It is found that these nonzero solutions describe states with a lower free energy than the zero solution when $T < T_C$. For $T > T_C$ an additional differentiation yields the small-signal dielectric constant:

$$\varepsilon^{-1}(E_{ex}) = \frac{\partial^2 F}{\partial D^2} \bigg|_{E=E_{ex}} = a(T - T_C) + 3bD(E_{ex})^2 + 5cD(E_{ex})^4$$

It is this quantity that is usually measured in e.g., capacitance experiments. For small values of E_{ex} we derive the following expression for the dielectric constant:

$$\varepsilon(E_{ex}) = \frac{1}{a(T - T_C)}$$

which is field-independent and exhibits a characteristic Curie-law behavior around T_C .

In experiments that involve moderate electric fields, the measured values of $\varepsilon(E_{ex})$ for paraelectrics ($T > T_C$) normally can be fitted to the following trend line: if $\varepsilon(E_{ex}) = \varepsilon_0 \kappa(E_{ex})$, then

$$\kappa(E_{ex}, T) \approx \kappa(0, T) - A(T)E_{ex}^2 + B(T)E_{ex}^4 + C(T)E_{ex}^6$$

Relating this to the Landau-Devonshire parameters is not straightforward, due to the implicit nature of equation 1. To deal with this problem, we expand the displacement D in powers of the external field: if $D = \varepsilon_0 \{\kappa_1 E + \kappa_3 E^3 + \kappa_5 E^5 + \kappa_7 E^7 \dots\}$, setting $a(T - T_C) \equiv \alpha$ gives

$$\begin{aligned}
E &= \alpha D + bD^3 + cD^5 \\
&= \alpha \varepsilon_0 \left(\kappa_1 E + \kappa_3 E^3 + \kappa_5 E^5 + \kappa_7 E^7 \dots \right) + b \varepsilon_0^3 \left(\kappa_1 E + \kappa_3 E^3 + \kappa_5 E^5 + \kappa_7 E^7 \dots \right)^3 \\
&\quad + c \varepsilon_0^5 \left(\kappa_1 E + \kappa_3 E^3 + \kappa_5 E^5 + \kappa_7 E^7 \dots \right)^5 \\
&= \alpha \varepsilon_0 \kappa_1 E + \left(\alpha \varepsilon_0 \kappa_3 + b \varepsilon_0^3 \kappa_1^3 \right) E^3 + \left(\alpha \varepsilon_0 \kappa_5 + 3b \varepsilon_0^3 \kappa_1^2 \kappa_3 + c \varepsilon_0^5 \kappa_1^5 \right) E^5 \\
&\quad + \left(\alpha \varepsilon_0 \kappa_7 + 3b \varepsilon_0^3 \kappa_1^2 \kappa_5 + 3b \varepsilon_0^3 \kappa_1 \kappa_3^2 + 5c \varepsilon_0^5 \kappa_3 \kappa_1^4 \right) E^7 \dots
\end{aligned}$$

The last term can be neglected since it is not needed to determine α , b , and c , so

$$E = \alpha \varepsilon_0 \kappa_1 E + \left(\alpha \varepsilon_0 \kappa_3 + b \varepsilon_0^3 \kappa_1^3 \right) E^3 + \left(\alpha \varepsilon_0 \kappa_5 + 3b \varepsilon_0^3 \kappa_1^2 \kappa_3 + c \varepsilon_0^5 \kappa_1^5 \right) E^5 + \dots$$

Equating the coefficients of like powers gives

$$\begin{aligned}
1 &= \alpha \varepsilon_0 \kappa_1 \\
0 &= \alpha \varepsilon_0 \kappa_3 + b \varepsilon_0^3 \kappa_1^3 \\
0 &= \alpha \varepsilon_0 \kappa_5 + 3b \varepsilon_0^3 \kappa_1^2 \kappa_3 + c \varepsilon_0^5 \kappa_1^5 \\
&\vdots \\
\Rightarrow \varepsilon_0 \kappa_1 &= \frac{1}{\alpha} \\
\varepsilon_0 \kappa_3 &= -\frac{b \left(\varepsilon_0 \kappa_1 \right)^3}{\alpha} = -\frac{b}{\alpha^4} \\
\varepsilon_0 \kappa_5 &= -\frac{3b \left(\varepsilon_0 \kappa_1 \right)^2 \varepsilon_0 \kappa_3 + c \left(\varepsilon_0 \kappa_1 \right)^5}{\alpha} = \frac{3b^2 - \alpha c}{\alpha^7}
\end{aligned}$$

This gives

$$D = \varepsilon_0 \kappa_1 \left\{ E_{ex} - b \left(\varepsilon_0 \kappa_1 \right)^3 E_{ex}^3 + \left(3b^2 \varepsilon_0 \kappa_1 - c \right) \left(\varepsilon_0 \kappa_1 \right)^5 E_{ex}^5 + \dots \right\}$$

and hence the dielectric constant

$$\varepsilon(E) = \frac{dD}{dE} = \varepsilon_0 \kappa_1 \left\{ 1 - 3b \left(\varepsilon_0 \kappa_1 \right)^3 E^2 + 5 \left(3b^2 \varepsilon_0 \kappa_1 - c \right) \left(\varepsilon_0 \kappa_1 \right)^5 E^4 + \dots \right\}$$

so that

$$\begin{aligned}\kappa(0,T) &= \kappa_1 \\ \varepsilon_0 A(T) &= 3b(\varepsilon_0 \kappa_1)^4 \\ \varepsilon_0 B(T) &= 5(3b^2 \varepsilon_0 \kappa_1 - c)(\varepsilon_0 \kappa_1)^6\end{aligned}$$

Note that even if the Landau-Devonshire parameters are temperature independent, the coefficients $A(T)$ and $B(T)$ will vary strongly with temperature around the Curie temperature due to the presence of high powers of κ_1 .

The coefficients $\varepsilon(0,T)$, $A(T)$, $B(T)$, and $C(T)$ can be obtained by fitting the experimental curves. A least-squares fit for the Curie temperature of the form

$$\alpha = a(T - T_C) = \frac{1}{\varepsilon_0 \kappa(0,T)},$$

where a and T_C are fitting parameters, is described in appendix. Once this is done, it is easy to derive the other Landau-Devonshire parameters:

$$b = \frac{1}{3} A(T) \varepsilon_0^{-3} [\kappa(0,T)]^{-4} \quad (2)$$

$$c = -\frac{1}{5} B(T) \varepsilon_0^{-5} [\kappa(0,T)]^{-6} + \frac{1}{3} A(T)^2 \varepsilon_0^{-5} [\kappa(0,T)]^{-7} \quad (3)$$

3. Fitting Results for BaTi_{0.9}(Sc,Ta)_{0.05}O₃

One effective way to modify the electrical properties of perovskite compounds like BaTiO₃ is to dope them with transition metals. We have found that this leads to materials that are both temperature insensitive and highly tunable, a circumstance that our group has investigated for several years. We use the following procedure to measure the dielectric constant versus temperature and field. Contacts are deposited by E-beam evaporation on circular samples of ~10.5 mm diameter and 0.5 mm thickness to form parallel-plate capacitors. The contacts are composed of layers of 250 Å Ti, 1500 Å Au, 3000 Å Ag, 1500 Å Au, in that order. The small-signal capacitance is measured by an impedance bridge in the temperature range $-55 \leq T \leq 120$ °C at 5 values of bias voltage across the capacitors from 0 to 500 V. The bias voltages needed for the measurement are provided by a Bertan 205B high-voltage D.C. power supply. The small-signal capacitance is given by the formula

$$C = \varepsilon_0 \chi_1 (E_{bias}) \frac{A}{h}$$

where A is the contact area, h is the sample thickness, and $E_{bias} = \frac{V_{bias}}{h}$, the electric field created within the sample by the bias voltage V_{bias} , was computed for each sample capacitor. This formula was inverted to give the dielectric response at each bias field.

It is a striking property of this material that, despite its chemical similarity to BaTiO_3 , it is unambiguously paraelectric over the entire temperature measurement range. In contrast, pure BaTiO_3 is ferroelectric over this range. A typical experimental curve of the dielectric constant versus field looks as seen figure 1.

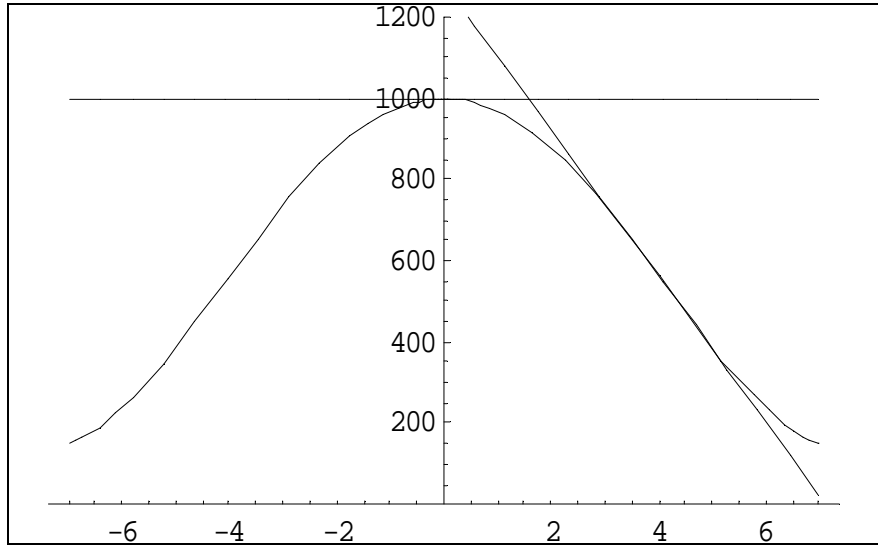


Figure 1. Typical experimental curve for ferroelectric dielectric constant.

The symmetry with field reflects the cubic nature of the crystal symmetry, which eliminates all odd powers of the electric field E . Comparing the shape of this curve with the expression

$$\varepsilon(E) = \varepsilon_1 \left\{ 1 - 3b\varepsilon_1^3 E^2 + 5(3b^2\varepsilon_1 - c)\varepsilon_1^5 E^4 + \dots \right\}$$

shows that it curves upward at very high fields, indicating that $3b^2\varepsilon_1 > c$ in our materials.

As stated above, our fitting procedure starts with the expression

$$\kappa(E_{ex}, T) \approx \kappa(0, T) - A(T)E_{ex}^2 + B(T)E_{ex}^4 + C(T)E_{ex}^6$$

where $\kappa(0, T)$ is taken directly from the zero-field measurement and $A(T)$, $B(T)$, and $C(T)$ are derived by least-squares fitting the difference

$$\begin{aligned} \Delta\kappa(E_{ex}, T) &= \kappa(E_{ex}, T) - \kappa(0, T) \\ &= -A(T)E_{ex}^2 + B(T)E_{ex}^4 + C(T)E_{ex}^6 \end{aligned}$$

Once this is done, the other Landau-Devonshire parameters are computed using equations 2 and 3. Figure 2 shows a typical curve fit of the experimental points (large dots) to this function at a specific temperature.

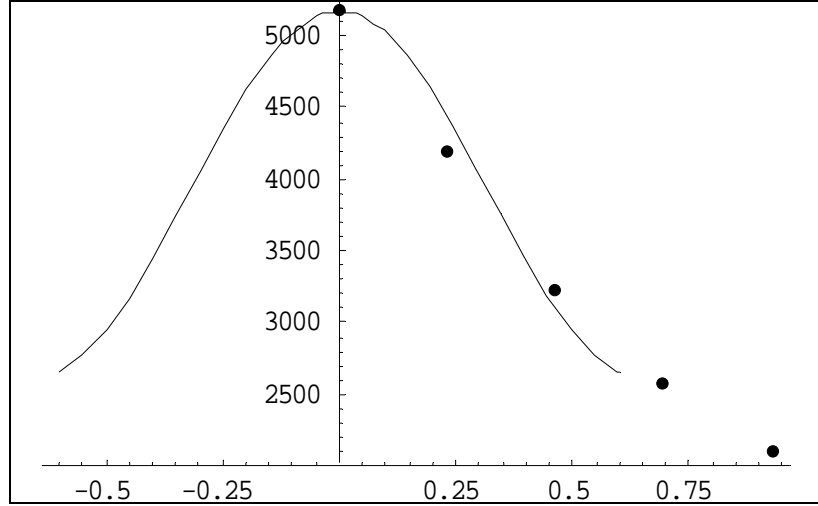


Figure 2. Theoretical fit for $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$ dielectric constant showing experimental data points.

The fitted values of a and T_C for this material are $4.2 \times 10^7 \frac{\text{V} - \text{m}}{\text{C} - ^\circ\text{K}}$ and -58°C . For comparison, the Curie constant $C = 1.8 \times 10^5 ^\circ\text{K}$ for single-crystal BaTiO_3 given in Lines and Glass (3), plus the transition temperature $T_C = 120^\circ\text{C}$, imply that $a = 3.7 \times 10^7 \frac{\text{V} - \text{m}}{\text{C} - ^\circ\text{K}}$ for pure BaTiO_3 (on the ferroelectric side of the Curie temperature). While the value of a for our material is reasonably close to this number, our fitted transition temperature for the (cubic-tetragonal) transition is remarkably low. Because pure BaTiO_3 undergoes three phase transitions, the highest (cubic-tetragonal) at around 120°C and the lowest (orthorhombic-rhombohedral) at -60°C , it is tempting to assume that in our material the two high-temperature phase transitions have been disrupted, leaving only the lowest-temperature one to form the observed peak in ϵ . This is in fact consistent with the behavior of PbTiO_3 . However, x-ray data indicate that the crystal structure of our material is cubic at room temperature, suggesting that the highest Curie temperature has indeed been moved down by this large amount.

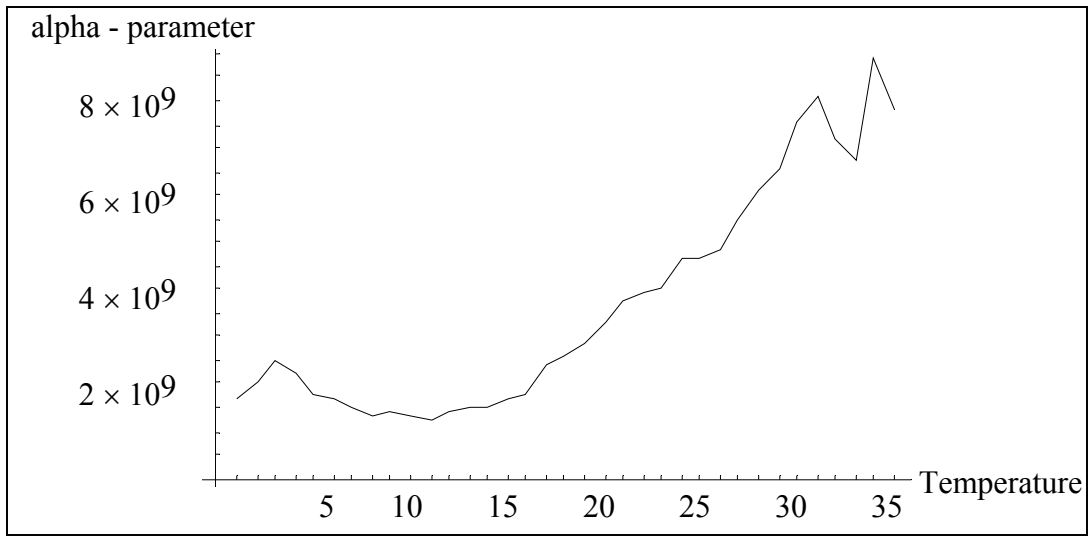


Figure 3. Fitted temperature variation of parameter α for $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$.

The other LD parameters b and c are strikingly different from those of pure BaTiO_3 .

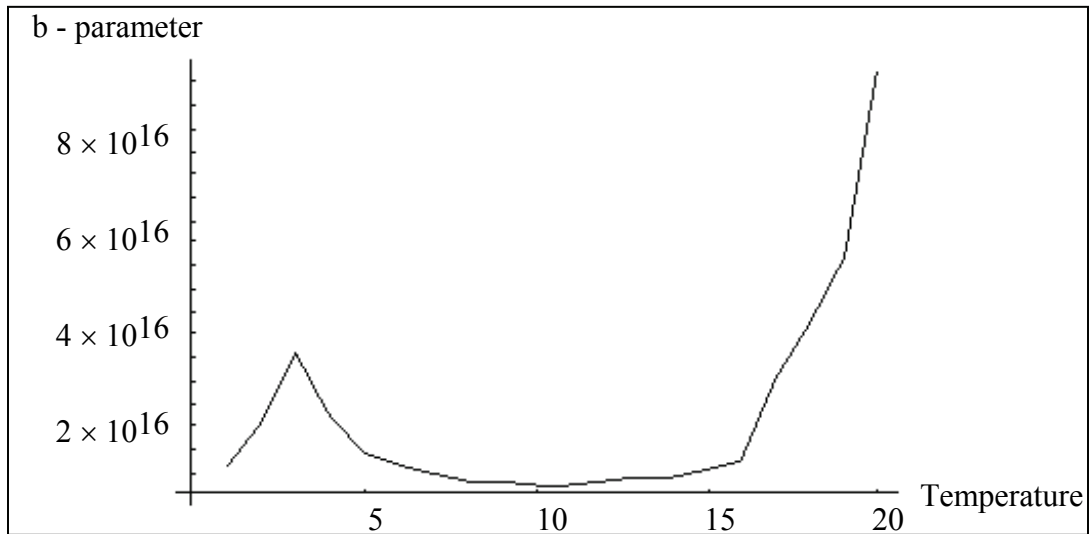


Figure 4. Fitted temperature variation of L.-D. parameter b for $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$.

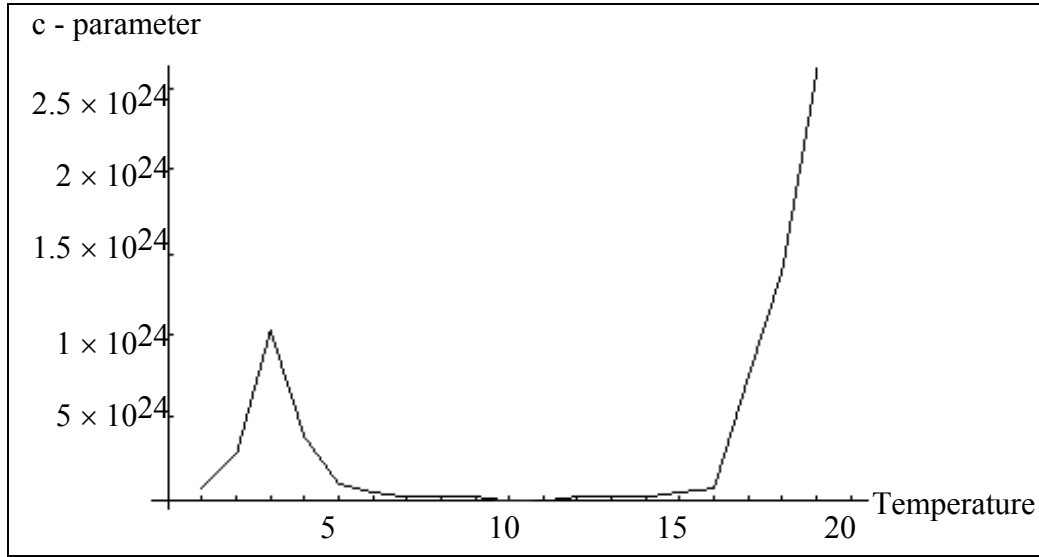


Figure 5. Fitted temperature variation of L.-D. parameter c for $\text{BaTi}_{0.9}(\text{Sc,Ta})_{0.05}\text{O}_3$.

Enumerating these differences, we have that

1. According to LD theory, the constant b should be negative for ferroelectrics with first-order phase transitions like BaTiO_3 . For our material this number is positive.
2. LD theory predicts that b and c should be temperature independent, whereas both are strongly temperature dependent in our material, although there is a range of temperatures where they are almost constant.
3. Both b and c are much larger than they are in BaTiO_3 . Their minimum values in the “flat”

regions of these curves (around room temperature) are $\sim 2 \times 10^{15} \frac{\text{V} \cdot \text{m}^5}{\text{C}^3}$ and

$\sim 8 \times 10^{21} \frac{\text{V} \cdot \text{m}^9}{\text{C}^5}$ respectively. Contrast these numbers with the values

$b = 6 \times 10^{13} \frac{\text{V} \cdot \text{m}^5}{\text{C}^3}$ and $c = 6 \times 10^{17} \frac{\text{V} \cdot \text{m}^9}{\text{C}^5}$ reported in (4).

One possible explanation for these anomalies is based on the commonly held idea that first order transitions in ferroelectrics are caused by electrostriction. When a system develops a spontaneous polarization, the associated internal field causes a spontaneous strain, which adds a term $-\frac{Q}{2Y}D^4$ to the free energy, where Q is the electrostrictive constant and Y is a combination

of elastic constants. Because this term is negative and proportional to D^4 , it reduces b in the free energy expression and can even make it negative, which converts the phase transition from second order to first order. Then the argument can be made that in BaTiO_3 the constant b is normally large, but is canceled out by this term, thereby converting a second-order phase

transition into the (weak) first-order transition observed in experiment. We tentatively advance the hypothesis that the elements we have added to the BaTiO_3 somehow eliminate the spontaneous strain, converting the crystal structure from tetragonal to cubic and “exposing” a large but previously hidden positive value of b . Similar arguments can be made for c . This hypothesis will be a subject for future research.

4. References

1. See, e.g.; Lines, M. E.; Glass, A. M. *Principles and Applications of Ferroelectrics and Related Materials*; Clarendon Press, Oxford, 1977, ch. 3.
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Appendix. Least-Squares Fit for the Curie Temperature

In calculating the fitting parameters it is convenient to use the inverse relation

$$\alpha = \frac{1}{\varepsilon(0, T)} = a(T - T_C)$$

Then the error function is

$$\Delta(a, T_C) = \sum_i \left[\alpha_i - a(T_i - T_C) \right]^2$$

where the summation is over the experimental data points. Minimizing this function with respect to a and T_C gives

$$\begin{aligned} \frac{\partial}{\partial a} \Delta(a, T_C) &= -\sum_i [T_i - T_C] 2 [\alpha_i - a(T_i - T_C)] = 0 \\ \frac{\partial}{\partial T_C} \Delta(a, T_C) &= \sum_i [a] 2 [\alpha_i - a(T_i - T_C)] = 0 \\ \Rightarrow \begin{cases} \sum_i \alpha_i [T_i - T_C] = a \sum_i [T_i - T_C]^2 \\ a \sum_i \alpha_i = a^2 \sum_i [T_i - T_C] \end{cases} \\ \Rightarrow \begin{cases} \sum_i \alpha_i T_i - T_C \sum_i \alpha_i = a \left(\sum_i T_i^2 - 2T_C \sum_i T_i + NT_C^2 \right) \\ \sum_i \alpha_i = a \sum_i [T_i - T_C] \Rightarrow a = \frac{\sum_i \alpha_i}{\sum_i T_i - NT_C} \end{cases} \end{aligned}$$

Let N be the number of data points, and

$$A = \frac{1}{N} \sum_i \alpha_i \quad B = \frac{1}{N} \sum_i T_i \quad C = \frac{1}{N} \sum_i T_i^2 \quad M = \frac{1}{N} \sum_i \alpha_i T_i$$

Then

$$\left. \begin{aligned} M - AT_C &= a(C - 2BT_C + T_C^2) \\ a &= \frac{A}{B - T_C} \end{aligned} \right\} \Rightarrow (B - T_C)(M - AT_C) = A(C - 2BT_C + T_C^2)$$

$$\Rightarrow BM - (M + AB)T_C + AT_C^2 = AC - 2ABT_C + AT_C^2$$

$$\Rightarrow BM - (M + AB)T_C = AC - 2ABT_C$$

$$\Rightarrow BM - AC = (M - AB)T_C$$

$$\Rightarrow T_C = \frac{BM - AC}{M - AB}$$

$$a = \frac{A}{B - \left\{ \frac{BM - AC}{M - AB} \right\}} = \frac{A(M - AB)}{B(M - AB) - (BM - AC)}$$

$$= \frac{A(M - AB)}{-AB^2 + AC} = \frac{M - AB}{C - B^2}$$

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